# 3P1-6

# Fabrication parameter effects on film quality in $Pb(Zr,Ti)O_3/Pb(Zr,Ti)O_3$

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## 1. Introduction

Speaker-based voice alert systems are being introduced to prevent accidents on motorways. One of these systems is the development of a highway alert speaker using ultrasonic waves. However, in conventional speaker manufacturing methods, the ultrasonic elements used in speakers consist of multiple components such as piezoelectric ceramics, metal plates and cone resonators in a resin case, making the manufacturing process complex and costly. Therefore, attention was turned to the sol-gel composite method[1,2)], which enables large-area application and reduced fabrication costs. The sol-gel composite piezoelectric films also have the advantage that they are flexible and do not interfere with vibrations even when the film thickness is large, as they have numerous pores.

From previous research, it was found that the speakers fabricated so far lacked sound pressure compared to existing mass-produced products, so one way to improve the sound pressure was to focus on improving the performance of the piezoelectric membrane. Therefore, one of the objectives was to reduce the porosity, as the piezoelectric strain constant $d_{33}$ decreases as the porosity increases. $Pb(Zr,Ti)O_3/Pb(Zr,Ti)O_3$ (PZT/PZT)[3,4)] was selected as the material because of its high piezoelectric strain constant and ease of polarisation, while HIZIRCO L was selected as the PZT piezoelectric powder because of its low dielectric constant and easy polarisation.

In this study, the effects on film thickness, permittivity, ultrasonic response and frequency response of PZT-L/PZT (L/PZT) prepared by mixing HIZIRCO L and PZT sol-gel solutions of PZT powder with a particle size of 1.2μm and 0.6μm in mixing ratios of 100:0, 75:25 and 50:50 were investigated.

## 2. Sample fabrication

First, HIZIRCO L and PZT sol-gel solutions mixed in three different mixing ratios were each mixed for 24 hours by a ball mill machine. The mixed sol-gel composite solution was applied to a titanium substrate 3 mm thick, 30 mm long and 30 mm wide using an automatic sprayer. Subsequently, drying was carried out for 5 min in the spray apparatus, followed by 10 min drying in a 150 °C baking furnace and 5 min annealing in a 650 °C baking furnace. The process from spray application to annealing was repeated five times. Poling by positive corona discharge was then carried out for 5 min. The DC power supply voltage used for poling was approximately 47 kV. The humidity during poling was kept below 20%. Finally, a top electrode with a diameter of 6 mm was fabricated by screen-printing. The samples fabricated with three different mixing ratios are shown in **Fig. 1**. No differences in the piezoelectric film surface were observed between the mixing ratios.

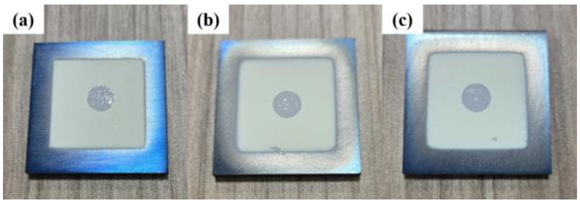


Fig. 1 Optical images of the sample for each mixing ratio (1.2μm:0.6μm) (a) 100:0 (b)75:25 (c) 50:50

## 2. Experiment method

Film thickness, relative permittivity ( $\varepsilon_r$ ), sensitivity of ultrasonic response and frequency response were used as evaluation indices. The film thickness was measured with a micrometer and the relative permittivity was calculated from the capacitance measured with an LCR meter. The ultrasonic response was measured by connecting a pulser/receiver (5073PR, OLYMPUS), oscilloscope and the prepared sample. The sensitivity of the ultrasonic response was calculated using Equation (1).

$$Sensitivity = -(20\log V_1/V_2 + P/R\ Gain) \quad (1)$$

The reference amplitude $V_1$ was set to 0.1 V and $V_2$ to $V_{P\text{-}P}$ of the first reflected wave from the bottom of the substrate. Finally, a Fast Fourier Transform (FFT) was performed on the same wave to measure the frequency response.

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## 4 Results and discussions

**Fig. 2** shows the ultrasonic response for each mixing ratio, while **Table I** shows the average and variance of film thickness, FFT results, relative permittivity and sensitivity for each mixing ratio. The average is the mean of three samples for each condition.

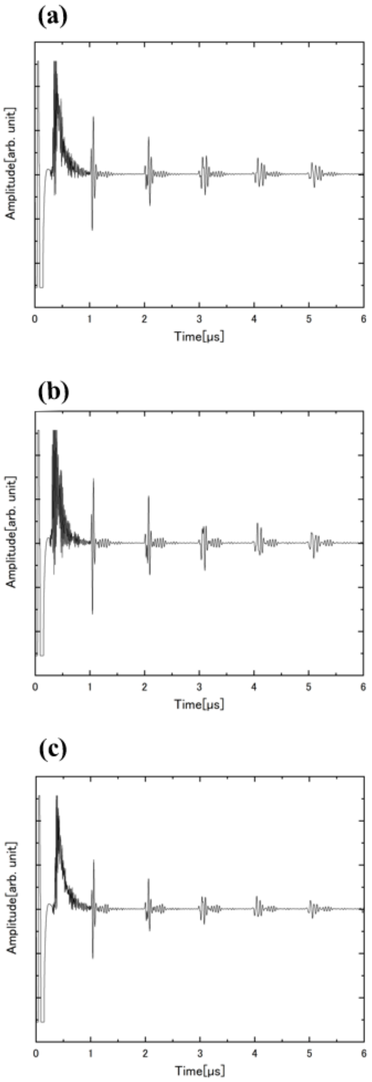


Fig.2 For each mixing ratio (1.2μm:0.6μm) the ultrasonic response (a)100:0 (b)75:25 (c)50:50

The film thickness was the largest for the mixing ratio 100:0. For the center frequency ($f_C$), the highest center frequency is found for the mixing ratio 75:25. For the relative permittivity, the mixing ratio 100:0 is the highest, but the dispersion variation of the mixing ratio 75:25 is small. From the variance of the relative permittivity and the center frequency, it can be considered that the porosity of mixing ratio 75:25 is the lowest because the film thickness of mixing ratio 75:25 is the lowest because the film thickness of mixing ratio 75:25 is the thinnest and the film thickness distribution is uniform. The reason why the relative permittivity is not high is that the thickness of films with poor film quality cannot be accurately measured with a micrometer. Therefore, the porosity of the 75:25 mixing ratio can be the smallest. Regarding sensitivity, the 50:50 mixing ratio has the highest sensitivity and the 75:25 mixing ratio has the lowest sensitivity. The reason for the lowest sensitivity at a mixing ratio of 75:25 is thought to be due to the large size of the upper electrode in relation to the film thickness and the electrical impedance mismatch.

Table I Results for each mixing ratio (1.2μm:0.6μm)

| Evaluation index | | mixing ration(1.2μm:0.6μm) | | |
|---|---|---|---|---|
| | | 100:0 | 75:25 | 50:50 |
| Film thickness[μm] | Ave. | 29 | 26 | 25 |
| | SD | 2.2 | 2.6 | 2.3 |
| $f_C$ [MHz] | Ave. | 21 | 25 | 22 |
| | SD | 2.5 | 0.0024 | 4 |
| -6dBBW | Ave. | 18 | 22 | 20 |
| | SD | 2.8 | 2.4 | 6.2 |
| $\varepsilon_r$ | Ave. | 300 | 280 | 250 |
| | SD | 530 | 90 | 240 |
| Sensitivity[dB] | Ave. | 6.0 | 5.9 | 9.3 |
| | SD | 0.11 | 0.48 | 1.3 |

## 4. Conclusion

In this study, L/PZT mixtures were prepared using PZT powder HIZIRCO L with particle sizes of 1.2μm and 0.6μm at ratios of 100:0, 75:25 and 50:50, and the film thickness, relative permittivity, sensitivity and frequency response were evaluated. It was confirmed that the dielectric constant with a mixing ratio of 75:25 had the highest center frequency with the least dispersion variation. This suggests that the porosity of the mixing ratio 75:25 is the lowest. However, the lower sensitivity of the 75:25 mixing ratio is an electrical impedance mismatch. Future work will focus on optimizing electrode size and increasing the number of experimental cycles.

## References

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